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## CAPILLARY AND CRYSTALIZATION STRENGTH OF GLASS BATCH COMPONENTS

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The attraction of glass particles simulating quartz sand grains for capillary rings of aqueous solutions of sodium sulfate and carbonate of various concentrations is experimentally investigated. It is shown that the attraction of particles at the first moment of contact with the bridging liquid is determined by the capillary effect. As water evaporates and the solution becomes saturated, additional cohesion of solid particles is registered, which is caused by the formation of crystallization contacts.

The moistening of the glass batch raises its thermal conductivity and decreases the entrainment of dust particles. The water menisci arising between solid particles cause capillary attraction and approach of the particles. The effect of the capillary attraction of particles is manifested when the powder contains 3–4% moisture [1]. In clotting, the drops of a moistening liquid transform the batch components into granules.

The capillary strength of the particle adhesion via concave liquid menisci is mostly determined by the Laplace pressure differential. The mobile liquid–air interface is deflected toward the phase of lower pressure. Quantitatively, the Laplace pressure differential  $\Delta P_L$  is calculated from the average meniscus curvature  $k_{av}$  and the surface tension  $\sigma$  of the liquid–gas interface.

$$\Delta P_L = \sigma k_{av} = \sigma \left( \frac{1}{\rho_2} - \frac{1}{\rho_1} \right),$$

where  $\rho_2$  and  $\rho_1$  are the main surface curvature radii at the selected point (Fig. 1).

In the general case, the capillary force of particle interaction  $f_{cap}$  generated by the drop liquid is made up of the Laplace pressure transmitted by the liquid via a section with area  $\omega = \pi a^2$  ( $a$  is the radius of the wetting parameter of a spherical particle) and the surface tension force applied to the perimeter  $L = 2\pi a$ :

$$f_{cap} = f_{cap}^v + f_{cap}^\omega = \Delta P \omega + \bar{\sigma} L, \quad (1)$$

where  $\bar{\sigma}$  is the projection of the surface tension vector applied to the three-phase boundary on the axis connecting the solid particles.

The direction of the first component  $f_{cap}^v$  depends on the mean curvature sign of the meniscus: at  $k_{av} > 0$  (a concave meniscus), the Laplace force draws the particles together, whereas at  $k_{av} < 0$  (a convex meniscus), the capillary pressure squeezes the particles out of the liquid. The second component  $f_{cap}^\omega$ , due to its physical meaning, is always positive.

In disclosing equation (1) via geometrical constructions, Fisher obtained the following formula:

$$f_{cap} = \sigma \pi R \sin \varphi \left[ R \sin \varphi \left( \frac{1}{\rho_2} - \frac{1}{\rho_1} \right) + 2 \sin(\varphi + \theta) \right], \quad (2)$$

where  $R$  is the spherical particle radius;  $\varphi$  is half of the central angle on which the liquid ring is resting;  $\theta$  is the wetting angle of the solid particle surface.

In the toroidal approximation, which approximates the meniscus by the surface of revolution of an arc with a constant curvature, the main curvature radii of the ring arising in

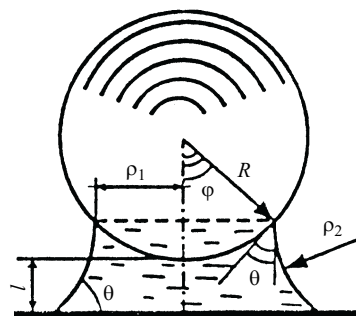
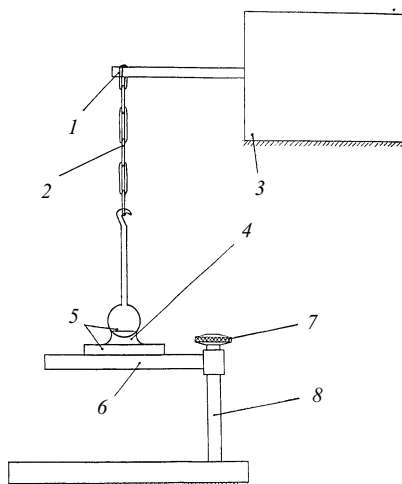


Fig. 1. Diagram of liquid interlayer connecting solid particles.

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**Fig. 2.** Diagram of the device for determining the capillary-crystallization strength of particles: 1) balance beam; 2) chain; 3) torsion balance; 4) capillary interlayer; 5) glass particles; 6) mobile support; 7) microscrew; 8) holder.

a sphere – plane contact are estimated from the following equations [2]:

$$\rho_1 = R \sin \varphi - \frac{l + R(1 - \cos \varphi)}{\cos(\varphi + \theta) + \cos \theta} [1 - \sin(\varphi + \theta)];$$

$$\rho_2 = \frac{l + R(1 - \cos \varphi)}{\cos(\varphi + \theta) + \cos \theta},$$

where  $l$  is the distance between the particles.

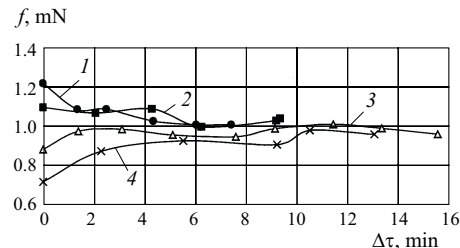
The reliability of the cohesion of solid batch particles fixed in a drop increases with increasing surface tension of the binding liquid and improved wetting (a decrease in the contact angle  $\theta$ ). When the ring has a small volume (angle  $\varphi \rightarrow 0$ ) and  $l = 0$ , Eq. (2) describing the contact between a spherical particle and a flat particle is transformed into the expression

$$f_{\text{cap}} = 4\sigma\pi R \cos \theta.$$

When a capillary cell is completely filled with liquid (angle  $\varphi \rightarrow 90^\circ$ ), the force decreases to

$$f_{\text{cap}} = 2\sigma\pi R \cos \theta.$$

The cohesion force in the present study was determined on a torsion balance. The diagram of the device is shown in Fig. 2. The cohesion forces were studied on a model simulating the capillary contact between a spherical particle, a liquid interlayer, and a flat substrate. The selected model particles were a glass ball 4.8 mm in diameter and a flat glass plate. The ball was produced by the fusion of a glass rod 1 mm in diameter. The spheroid particle was suspended on a balance beam, and the flat particle was brought from underneath using a microscrew installed on a holder. Distilled water or



**Fig. 3.** Dependence of capillary force on contact duration (ring material: distilled water, spherical particle diameter 4.8 mm). Weight of liquid on a spherical particle at the initial moment: 1, 2, 3, and 4) 0.007250, 0.011650, 0.013700, and 0.019100 g, respectively.

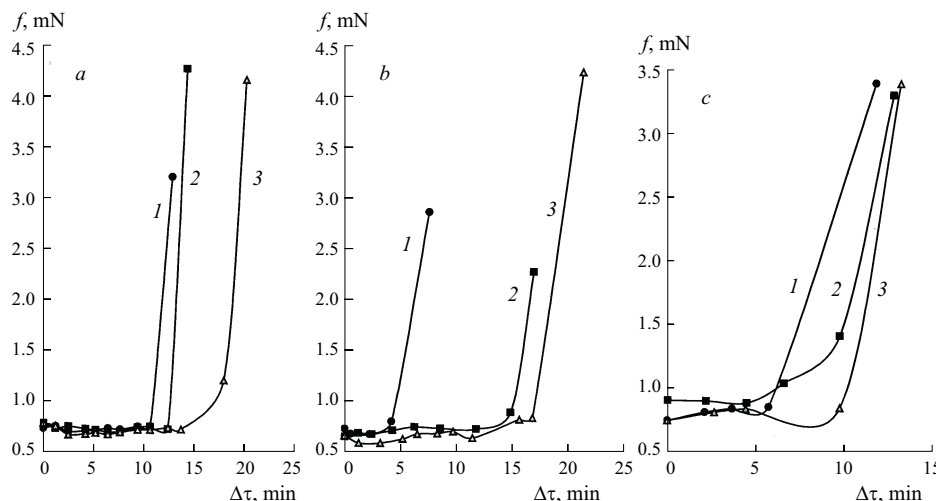
aqueous solutions of sodium carbonate and sulfate serves as capillary liquid.

The model glass particles were used in the experiment to study the capillary and crystallization strength in the course of the ring liquid evaporation. The salts selected for the experiments are present in the glass batch, and their solutions moisten well the solid particles and form capillary contacts. A drop of water or the solution of a preset concentration was deposited on the lower surface of the spherical particle. The initial weight of the capillary meniscus was determined on an analytical balance before and after the deposition of the liquid.

In the case of good wettability, a concave liquid ring is formed, which attracts particles and draws them to a direct contact. The capillary force was determined on taking off the lower particle. When the external force caused by the deformation of the torsion balance spring exceeds the maximum capillary force, the capillary neck is ruptured. The capillary force was registered both at the moment of ring formation and after preset time periods. The experiment was repeated several times. The obtained experimental data were used to plot the dependence of the capillary force (for distilled water) and of the capillary-crystallization strength (for salt solutions) depending on the contact duration.

In wetting a batch with distilled water, the liquid drops penetrate between the insoluble quartz sand grains and draw them together without modifying their composition. Therefore, a pure capillary effect is observed in water (Fig. 3). In a sphere – plane contact, the capillary strength determined by the water meniscus varies from  $4$  to  $2\sigma\pi R$  ( $R$  is the sphere radius). The boundary values of the capillary force calculated for a ball 4.8 mm in diameter according to the above-mentioned expressions constitute 2.17 and 1.08 mN, respectively.

In the initial period of a capillary contact between particles, the force value is inverse to the liquid weight. After an exposure, the attraction force becomes equal for all initial volumes. After 10 min this force is equal to 0.95 mN, which correlates with the surface tension  $63 \text{ mJ/m}^2$ . The analysis of the experimental curves indicates that the force variation in a small volume of liquid is determined by an increased concentration of surfactant impurities. In large quantities of capillary liquid, the force increases mainly due to a decreased volume of the liquid ring. The evaporation duration is regu-



**Fig. 4.** Dependence of capillary force on contact duration (ring material: soda solution, spherical particle diameter 4.8 mm): *a*) solution concentration 0.50 M, weight of liquid on a spherical particle at the initial moment: 1, 2, and 3) 0.004218, 0.008100, and 0.009350 g, respectively; *b*) solution concentration 1.00 M, weight of liquid on a spherical particle at the initial moment: 1, 2, and 3) 0.002650, 0.006300, and 0.006300 g, respectively; *c*) solution concentration 2.00 M, weight of liquid on a spherical particle at the initial moment: 1, 2, and 3) 0.006850, 0.009050, and 0.009400 g, respectively.

lated by the liquid volume, which is generally corroborated by the experimental curves.

The dissolution of salts improves the wettability of sand grains and facilitates their being enveloped in a thin film of salt. At the same time, the contact between the components improves, which accelerates the silicate formation reactions. The dissolution of salt in an aqueous capillary interlayer qualitatively modifies the nature of the reactions between the solid particles.

In the case of using salt solutions, after a certain time, crystallization of salt from the saturated solution was observed as a result of water evaporation from the capillary meniscus. The evaporation originated from the outer layers of the liquid ring along the wetting perimeter, therefore, after the removal of particles, salt crystal deposition in a circular shape was visible.

After a certain critical time passes, the crystalline depositions started merging, growing both from the flat particle and from the spherical particle.

The crystallization strength at first complements the capillary force and after the liquid evaporates, it fully controls the strength of the contact. The crystallization strength exceeds many times the capillary attraction of particles, occasionally by an order of magnitude. In general, the arrangement of curves on the capillary segment of the dependence  $f=f(\tau)$  corresponds to the earlier established regularity: as the liquid volume increases, the capillary force decreases.

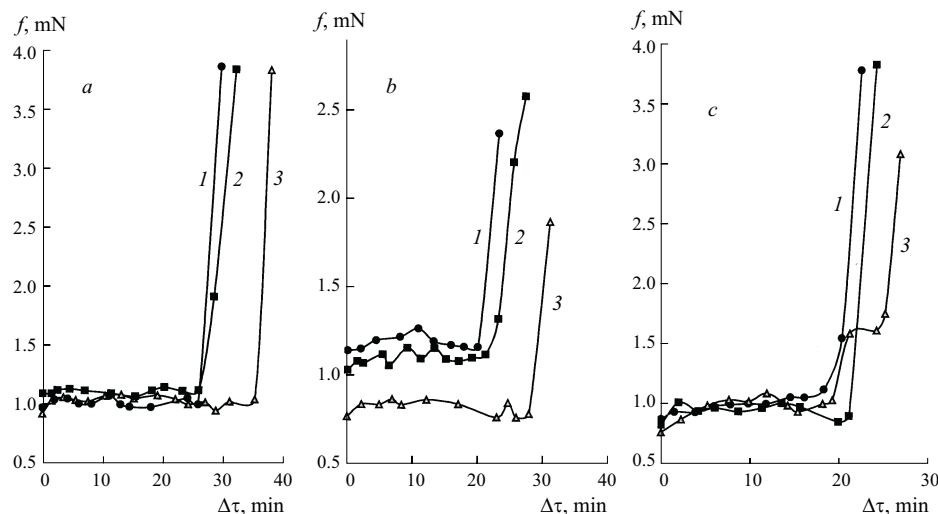
For a low-concentration solution (0.50 M), the capillary cohesion after 13–17 min is transformed into crystallization cohesion (Fig. 4*a*). The evaporation rate under low concentrations is relatively low; therefore, the cohesion strength could be measured 3–4 times before the moment at which

the strength exceeded the measurement limit of the balance. Generally, the capillary force after 17 min is completely transformed into crystallization strength.

When the concentration of the solution is doubled (1.00 M), the extent of the capillary segment increases (Fig. 4*b*). On the whole, the capillary strength value varies within the same interval as it does under low concentrations, but as the contact duration increases, a growth in strength is registered. Apparently, the capillary force already at the early stages of evaporation coincides with the crystallization cohesion of particles.

In high-concentration solutions (2.00 M), the pure capillary interaction of particles is virtually absent (Fig. 4*c*). The crystallization bonds between the sphere and the plate cause a significant increase in the contact strength already during the first minutes, the evaporation is much more intense than in the case of water, and after 9–10 min the liquid ring totally disappears. In the case of water, evaporation lasted 20–25 min.

The maximum concentration in determining the strength of contact in sodium sulfate solutions was 1 M, since this solution becomes oversaturated at lower concentrations. When the solution concentration is minimum (0.25 M), the capillary segment of the curve correlates with the strength 0.9–1.1 mN. The regular increase in capillary strength, as the volume decreases, is observed in this case, i.e., curve 2 is arranged below curves 1 and 3 (Fig. 5*a*). After a 25 min contact, the shape of the curves is determined by the crystallization strength, i.e., the sedimentation of sodium sulfate crystals. The main regularities of the strength of the contact implemented via an interlayer of sodium sulfate solution are the



**Fig. 5.** Dependence of capillary force on contact duration (ring material: sodium sulfate solution, spherical particle diameter 4.8 mm): *a*) solution concentration 0.25 M, weight of liquid on a spherical particle at the initial moment: 1, 2, and 3) 0.005050, 0.006230, and 0.007050 g, respectively; *b*) solution concentration 0.50 M, weight of liquid on a spherical particle at the initial moment: 1, 2, and 3) 0.004200, 0.006200, and 0.008150 g, respectively; *c*) solution concentration 1.00 M, weight of liquid on a spherical particle at the initial moment: 1, 2, and 3) 0.007150, 0.008200, and 0.009900 g, respectively.

same as those established for the contact implemented via soda solution.

The capillary segment of the curve for sodium sulfate solution with concentration 0.50 M (Fig. 5*b*) is somewhat shorter, and the curves are shifted further apart depending on the liquid volume: the greater the volume of the liquid, the lower is the curve position, i.e., the lower is the capillary force. In the cases where the difference in volumes is significant, the curves are far apart. At the maximum sodium sulfate concentration, the time of existence of the capillary neck is shorter by approximately 2 – 4 min (Fig. 5*c*).

Thus, the moistening of glass batch produces fixation of particles via liquid menisci. The capillary interlayer of car-

bonate and sulfate solutions cause the attraction of glass batch particles both at the stage of capillary cohesion and at the stage of formation of crystallization contacts. Accordingly, glass batch moistening redistributes the soluble components, transferring them via dissolution – crystallization processes to the contact areas of sand grains.

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